

# Mass Measurements and Implications for the Energy of the High-Spin Isomer in $^{94}\text{Ag}$

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Nuclides in the vicinity of  $^{94}\text{Ag}$  have been studied with the Penning trap mass spectrometer JYFLTRAP at the Ion-Guide Isotope Separator On-Line. The masses of the two-proton-decay daughter  $^{92}\text{Rh}$  and the beta-decay daughter  $^{94}\text{Pd}$  of the high-spin isomer in  $^{94}\text{Ag}$  have been measured, and the masses of  $^{93}\text{Pd}$  and  $^{94}\text{Ag}$  have been deduced. When combined with the data from the one-proton- or two-proton-decay experiments, the results lead to contradictory mass excess values for the high-spin isomer in  $^{94}\text{Ag}$ ,  $-46370(170)$  or  $-44970(100)$  keV, corresponding to excitation energies of  $6960(400)$  or  $8360(370)$  keV, respectively.

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Exotic decay modes of the highest-spin isomer in the  $N = Z$  nuclide  $^{94}\text{Ag}$  have been puzzling nuclear physicists for several years. The properties of this isomer are unprecedented in the entire known Segré chart and have resulted in a series of intensive studies [1, 2, 3, 4, 5, 6, 7]. Because of its high excitation energy, this isomer with a half-life of  $T_{1/2} = 0.39(4)$  s [4] and spin ( $21^+$ ), can decay via  $\beta$  decay or  $\beta$ -delayed proton emission up to three protons or directly via one- or two-proton decay. Recently, the possibility of the two-proton decay mode was questioned [7]. In order to uncover the nature of this isomer and its possible decay modes, the decay energies, i.e., the masses of the nuclei involved, should be experimentally determined, since they are based on extrapolations of systematic trends in the Atomic Mass Evaluation 2003 (AME) [8]. In this Letter, we present the results from precision mass measurements around  $^{94}\text{Ag}$  and discuss their impact on the high-spin isomer in  $^{94}\text{Ag}$ .

The masses of  $^{84}\text{Y}$ ,  $^{87}\text{Zr}$ ,  $^{88,89}\text{Mo}$ ,  $^{88-92}\text{Tc}$ ,  $^{90-94}\text{Ru}$ ,  $^{92-95}\text{Rh}$ , and  $^{94-96}\text{Pd}$  have been measured with JYFLTRAP [9], a double Penning trap at the IGISOL (Ion-Guide Isotope Separator On-Line) [10], in a joint project with SHIPTRAP ion trap facility at GSI [11, 12] to investigate nuclides in the region of the  $rp$ - and possible  $\nu p$ -process paths. The results of the project will be published in a separate paper [13]. Of the measured nuclides, the masses of the two-proton-decay daughter  $^{92}\text{Rh}$  and the beta-decay daughter  $^{94}\text{Pd}$  given in Table I are essential for the study of  $^{94}\text{Ag}$ . These nu-

clides were produced via heavy-ion fusion-evaporation reactions induced by a  $^{40}\text{Ca}$  beam on a  $^{nat}\text{Ni}$  target at the IGISOL where almost all reaction products end up at a  $q = 1^+$  charge state. The ions were accelerated to 30 keV, mass-separated and delivered to a radio frequency quadrupole (RFQ) ion beam cooler and buncher [14]. The RFQ transferred the ion bunches to JYFLTRAP which consists of two Penning traps situated in a homogeneous magnetic field. The first trap is used for isobaric purification by mass-selective buffer-gas cooling [15] and the second trap is dedicated to high-precision mass measurements via a time-of-flight cyclotron frequency determination [16]. By measuring the time of flight as a function of excitation frequency, the cyclotron frequency  $\nu_c = qB/(2\pi m)$  can be determined. The magnetic field  $B$  is calibrated by measuring well-known reference masses, for example, the nuclides  $^{85}\text{Rb}$  and  $^{94}\text{Mo}$  in this work. From the measured cyclotron frequency ratios  $\nu_{c,ref}/\nu_c$ , the mass excess values  $\Delta$  were derived. With this method, a typical mass uncertainty below 10 keV was achieved for the nuclides.

Although the mass of  $^{94}\text{Ag}$  was not directly measured, the mass of its  $\beta$ -decay daughter,  $^{94}\text{Pd}$ , was determined in this work. Since the ground state of  $^{94}\text{Ag}$  is presumably a  $T = 1$  isobaric analog state [17], the  $Q_{EC}$  value of  $^{94}\text{Ag}$  can be estimated quite accurately from the Coulomb displacement energy  $\Delta E_C = Q_{EC} + \Delta_{nH}$ , where  $\Delta_{nH}$  is the neutron-hydrogen mass difference [8]. Thus, the mass of  $^{94}\text{Ag}$  can be obtained from the  $Q_{EC}$  value and the mass of  $^{94}\text{Pd}$ . Experimental Coulomb displacement energies have been investigated as a function of  $Z_{\text{average}}/A^{1/3}$  in Ref. [18], where  $Z_{\text{average}} = \frac{1}{2}(Z_{\text{mother}} + Z_{\text{daughter}})$ . However, the linear fit results in a significant deviation for the heaviest known  $T = 1$  nucleus  $^{74}\text{Rb}$ . Therefore, a new fit to the Coulomb displacement energies based on the current experimental  $Q_{EC}$  values of the odd-odd  $N = Z$  nuclei is shown in Fig. 1. This new fit includes nuclides such as  $^{62}\text{Ga}$  [19],  $^{66}\text{As}$  [20] and  $^{74}\text{Rb}$

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TABLE I: Mass excess values  $\Delta$  for  $^{92}\text{Rh}$  and  $^{94}\text{Pd}$ . The mass excess values were derived using the mass of a reference nuclide as given in Ref. [8]. In column four, “#” indicates a value that is derived from experimental, systematic trends [8].

Nuclide	Ref. atom	$\Delta_{\text{JYFL}}$ (keV)	$\Delta_{\text{AME}}$ (keV)	$\Delta_{\text{JYFL-AME}}$ (keV)
$^{92}\text{Rh}$	$^{85}\text{Rb}$	$-62998.6(4.3)^a$	$-63360(400)\#$	$361(400)$
$^{94}\text{Pd}$	$^{94}\text{Mo}$	$-66097.9(4.7)$	$-66350(400)\#$	$252(400)$

<sup>a</sup>An average of the JYFLTRAP and SHIPTRAP values.

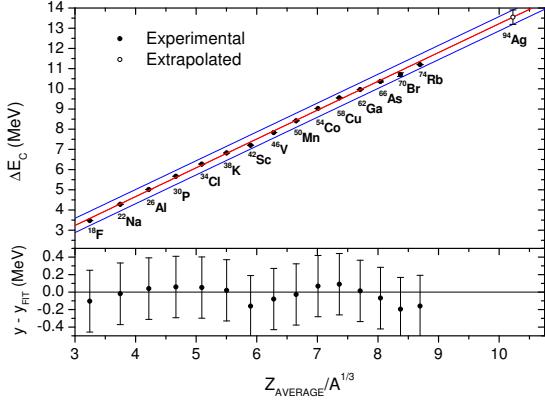


FIG. 1: (color online). Experimental Coulomb displacement energies for odd-odd  $N = Z$  nuclei. A 68 % prediction band gives  $\Delta E_C = 13550(360)$  keV for  $^{94}\text{Ag}$ . The  $Q_{\text{EC}}$  values of  $^{26}\text{Al}$  [29],  $^{42}\text{Sc}$  [29],  $^{46}\text{V}$  [29],  $^{50}\text{Mn}$  [30],  $^{54}\text{Co}$  [30],  $^{62}\text{Ga}$  [19], and  $^{74}\text{Rb}$  [21] as well as the masses of  $^{22}\text{Na}$  [31],  $^{38}\text{K}$  [32] and  $^{66}\text{As}$  [20] are from recent Penning trap measurements. The value for  $^{34}\text{Cl}$  is taken from the compilation of Hardy and Towner [33] and for  $^{70}\text{Br}$  from Refs. [33, 34]. All other values are from Ref. [8]. The residuals of the fit together with the errors representing the 68 % prediction band are shown in the lower panel.

[21] for which precise Penning trap measurements are now available. With a 68 % prediction band, the fit gives  $\Delta E_C = 13550(360)$  keV and  $Q_{\text{EC}} = 12760(360)$  keV in agreement with the  $Q_{\text{EC}}$  values based on the half-life [22] and on the systematics [8] (see Table II). The values from the Coulomb displacement energy formula for  $T = 1$  in [18],  $\Delta E_C = 13552(3)$  keV and  $Q_{\text{EC}} = 12770(3)$  keV, agree perfectly with the new fit.

From the  $Q_{\text{EC}}$  value and the mass of  $^{94}\text{Pd}$ , a mass excess of  $-5330(360)$  keV is obtained for  $^{94}\text{Ag}$ . From the masses of  $^{94}\text{Ag}$  and  $^{92}\text{Rh}$ , a two-proton separation energy in  $^{94}\text{Ag}$  is calculated,  $S_{2p} = -\Delta(^{94}\text{Ag}) + \Delta(^{92}\text{Rh}) + 2\Delta(^1\text{H}) = 4910(360)$  keV. If this two-proton separation energy is combined with the two-proton-decay data of the high-spin isomer in  $^{94}\text{Ag}$  [ $E_{2p} = 1900(100)$  keV [6] and  $E_x(^{92}\text{Rh}) = 1548.6(14)$  keV [23]], an excitation energy  $E_x = 8360(370)$  keV is obtained for this isomeric state. When combined with the measured  $^{92}\text{Rh}$  mass, the two-proton-decay  $Q$  value [6] gives a mass excess  $\Delta = -44970(100)$  keV for the high-spin isomer. The newly derived values differ significantly from the values based on the systematics [ $E_x = 6670(640)\#$  keV [8] and  $\Delta = -46800(500)\#$  keV [8]] and the empiri-

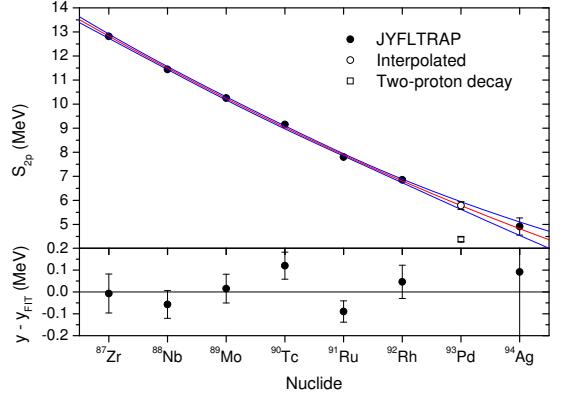


FIG. 2: (color online). Two-proton separation energies for the  $N = 47$  isotones measured at JYFLTRAP. A parabolic fit shown as the red line gives  $S_{2p} = 5780(160)$  keV for  $^{93}\text{Pd}$ . The  $S_{2p}(^{93}\text{Pd}) = -\Delta(^{93}\text{Pd}) + \Delta(^{91}\text{Ru}) + 2\Delta(^1\text{H}) = 4380(100)$  keV using the  $\Delta(^{93}\text{Pd}) = -S_p(^{93}\text{Pd}) + \Delta(^{92}\text{Rh}) + \Delta(^1\text{H})$  with  $S_p(^{93}\text{Pd}) = 2330(100)$  keV [6] and the measured masses of  $^{91}\text{Ru}$  and  $^{92}\text{Rh}$  shown as an open square clearly deviates from the trend. The residuals of the fit are shown in the lower panel.

cal shell model [ $E_x = 6300$  keV [3]]. However, the excitation energy is within the extrapolation of  $E_x = 6500(2000)\#$  keV in Ref. [24].

The difference in the isomeric energy between this work and [5] could be due to the extrapolated proton separation energy of  $^{94}\text{Ag}$  [ $S_p = -\Delta(^{94}\text{Ag}) + \Delta(^{93}\text{Pd}) + \Delta(^1\text{H}) = 890(640)\#$  keV [8]] which was used together with the proton-decay data [ $Q_p = \Delta(^{94}\text{Ag}(21^+)) - \Delta(^{93}\text{Pd}) - \Delta(^1\text{H}) = 5780(30)$  keV [5]] to determine the excitation energy. In order to study this, the mass of  $^{93}\text{Pd}$  was derived from an interpolation of the two-proton separation energies in the  $N = 47$  isotones shown in Fig. 2. Here the masses from zirconium to rhodium were determined at JYFLTRAP [13, 25] and the values for  $^{85}\text{Sr}$  and  $^{86}\text{Y}$  are from Ref. [8]. A parabolic fit yields  $S_{2p}(^{93}\text{Pd}) = 5780(160)$  keV, which results in a mass excess of  $\Delta(^{93}\text{Pd}) = -S_{2p}(^{93}\text{Pd}) + \Delta(^{91}\text{Ru}) + 2\Delta(^1\text{H}) = -59440(160)$  keV and  $S_p(^{93}\text{Pd}) = -\Delta(^{93}\text{Pd}) + \Delta(^{92}\text{Rh}) + \Delta(^1\text{H}) = 3730(160)$  keV.

The mass of  $^{93}\text{Pd}$  yields a proton separation energy of  $1180(390)$  keV for  $^{94}\text{Ag}$ . If we now combine the one-proton decay data [5] with this proton separation energy and the mass of  $^{93}\text{Pd}$ , an excitation energy of  $6960(400)$  keV and a mass excess of  $-46370(170)$  keV is obtained for the  $(21^+)$  isomer in agreement with the ex-

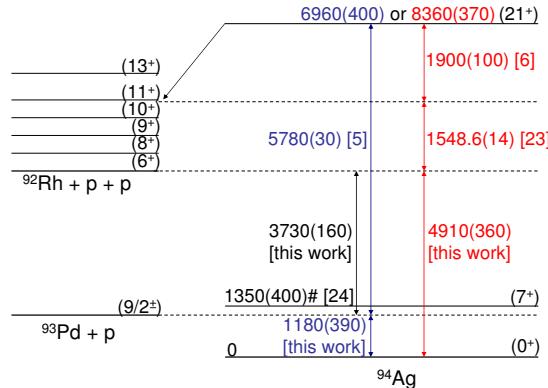


FIG. 3: (color online). Decay scheme of  $^{94}\text{Ag}$  ( $21^+$ ). The excitation energy of  $^{94}\text{Ag}$  ( $21^+$ ) depends on whether it is determined from the two-proton separation energy of  $^{94}\text{Ag}$  and the two-proton-decay data [6] (shown in red) or from the proton separation energy of  $^{94}\text{Ag}$  and the proton-decay  $Q$  value [5] (shown in blue). The energies are given in keV.

citation energy given in Ref. [5]. Thus, the uncertainty in the extrapolated proton separation energy in Ref. [8] does not explain the observed difference in the excitation energy of the high-spin isomer. The two-proton-decay data lead to a significantly different excitation energy than the one-proton-decay data as shown in Fig. 3. A summary and the results for  $^{94}\text{Ag}$  and  $^{93}\text{Pd}$  are given in Table II and compared with the values from the AME 2003 [8].

In order to explain the discrepancy in the excitation energy of  $^{94}\text{Ag}$  ( $21^+$ ), either the one-proton-decay  $Q$  value is too low or the two-proton-decay energy is too high. In a recent paper [7], it was claimed that the two-proton decay of  $^{94}\text{Ag}$  ( $21^+$ ) to the 1549-keV state in  $^{92}\text{Rh}$  would be highly unlikely, since the most probable state to be populated has an excitation energy of 240(570) keV [7]. With the revised masses of  $^{92}\text{Rh}$ ,  $^{93}\text{Pd}$ , and  $^{94}\text{Ag}$  ( $21^+$ ) determined in this work, the most probable level to be fed lies at 150(190) keV when based on the one-proton-decay data and agrees with the 1549-keV level in  $^{92}\text{Rh}$  when based on the two-proton-decay data.

The only way to energetically enable the two-proton decay is if the excitation energy of the ( $21^+$ ) isomer in  $^{94}\text{Ag}$  lies at 8360(370) keV instead of 6960(400) keV. A sizable increase in the proton-decay  $Q$  value is required to explain the 1400(540) keV difference between the excitation energies. This could be possible if the proton decay fed higher-lying states than the 4994-keV and 4751-keV states observed in Ref. [5]. For example, the 653-keV  $\gamma$  rays following the de-excitations of the 5648-keV ( $37/2^+$ ) and 6994-keV ( $39/2^+$ ) states [26] may have been hidden by a large background of  $\beta$ -delayed  $\gamma$  rays in Ref. [5]. Another possibility is that some transitions have been missed in the decay scheme of  $^{93}\text{Pd}$  in earlier experiments.

As the  $T = 1$  state is not always the lowest state in a  $T_Z = 0$  nucleus (for example, in  $^{58}\text{Cu}$ ), it is worthwhile to consider whether the mass calculated from the

TABLE II: Deduced results for  $^{94}\text{Ag}$  and  $^{93}\text{Pd}$  in comparison with the AME 2003 values based on systematics (#) [8].

	JYFL (keV)	AME (keV)	JYFL–AME (keV)
$^{94}\text{Ag}$	$\Delta$ $-53330(360)$	$-53300(500)\#$	$-30(620)$
	$Q_{\text{EC}}$ $12760(360)$	$13050(640)\#$	$-290(740)$
	$S_p$ $1180(390)$	$890(640)\#$	$290(760)$
	$S_{2p}$ $4910(360)$	$4520(640)\#$	$400(740)$
$^{93}\text{Pd}$	$\Delta$ $-59440(160)$	$-59700(400)\#$	$260(440)$
	$S_p$ $3730(160)$	$3630(570)\#$	$100(590)$
	$S_{2p}$ $5780(160)$	$5620(710)\#$	$160(730)$

Coulomb displacement energies for  $^{94}\text{Ag}$  is a ground-state mass. For example, a ( $7^+$ ) isomeric state is expected at 661 [2] or 1350(400)#[24] keV in  $^{94}\text{Ag}$ . If the ground state of  $^{94}\text{Ag}$  were the  $7^+$  state, this would result in a lower ground-state mass, a higher two-proton separation energy, and, hence, an even higher excitation energy of the high-spin ( $21^+$ ) isomer. In contradiction to this latter consideration, the ground state is considered to be a  $T = 1$  state [17].

The excitation energy of 8360(370) keV for the high-spin isomer lies above the suggested  $T = 1$ , ( $20^+$ ) isobaric analogue state (IAS) which has been experimentally observed at 7.7 MeV in  $^{94}\text{Pd}$  [3] and should lie at the same excitation energy in  $^{94}\text{Ag}$ . Although the higher excitation energy enables a two-proton decay, it is difficult to explain why this ( $21^+$ ) state is isomeric and does not rapidly decay to the ( $20^+$ ) state. One explanation is that, due to unobserved or wrongly assigned  $\beta$ -delayed  $\gamma$  rays from the  $^{94}\text{Ag}$  ( $21^+$ ) decay, the observed ( $20^+$ ) level in  $^{94}\text{Pd}$  may in reality be a  $18^+$  state which would mean that the ( $20^+$ ),  $T = 1$  state would lie higher in both  $^{94}\text{Pd}$  and  $^{94}\text{Ag}$ . The high-spin isomer has been suggested to be highly deformed in Ref. [6] but this has later been questioned by the calculations in Refs. [7, 27]. If the ( $20^+$ ) IAS lies below the ( $21^+$ ) high-spin isomer and a possible high deformation of this isomer does not explain the hindrance of the internal decay, the excitation energy of the isomer has to be lower, supporting the one-proton-decay data.

In conclusion, one-proton-decay data [5] and two-proton-decay data [6] disagree with each other when the mass excess values for  $^{92}\text{Rh}$ ,  $^{93}\text{Pd}$  and  $^{94}\text{Ag}$  are combined with these data. A possible explanation for this discrepancy is that some  $\gamma$  transitions have not been observed in the one-proton decay due to a large background of  $\beta$ -delayed  $\gamma$  rays. This would suggest that the excitation energy of the  $^{94}\text{Ag}$  ( $21^+$ ) isomer lies at around 8.4 MeV instead of 7.0 MeV. Although the excitation energy of the high-spin isomer in  $^{94}\text{Ag}$  remains uncertain, we have obtained new data on its decay  $Q$  values tabulated for the two different excitation energies obtained in this work in Table III. In addition, we have considerably reduced the uncertainties of the one-, two- and three-proton separation energies in  $^{94}\text{Pd}$  needed in the decay studies of  $^{94}\text{Ag}$  ( $21^+$ ). Further experiments on the one-proton decay of

TABLE III: Results for the high-spin ( $21^+$ ) isomer in  $^{94}\text{Ag}$  (given in keV) in comparison with the values from the AME 2003 [8] based on systematics (#). When combined with the data obtained in this work, one-proton- [5] and two-proton-decay [6] data lead to different results.

$^{94}\text{Ag}$ ( $21^+$ )	One-proton [5]	Two-proton [6]	AME 2003
$\Delta$	-46370(170)	-44970(100)	-46800(500) #
$E_x$	6960(400)	8360(370)	6500(2000) # [24]
$Q_{\text{EC}}$	19720(170)	21130(100)	19550(640) #
$Q_p$	5780(30) [5]	7180(190)	5610(640) #
$Q_{2p}$	2050(170)	3450(100) [6]	1980(640) #

$^{94}\text{Ag}$  ( $21^+$ ) and on the decay schemes of  $^{92}\text{Rh}$  and  $^{93}\text{Pd}$  could verify the proton-decay  $Q$  value. To finally solve this puzzle, direct mass measurements on  $^{93}\text{Pd}$ ,  $^{94}\text{Ag}$  and  $^{94}\text{Ag}$  ( $21^+$ ) are needed, posing new challenges for the production of these exotic species. These challenges are presently being pursued at the IGISOL facility [28].

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